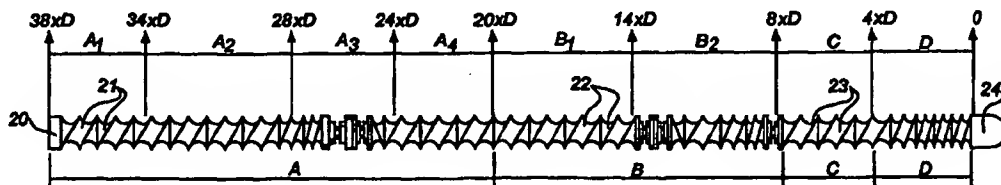




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(54) Title: PROCESS FOR CONTINUOUSLY MANUFACTURING COMPOSITES OF POLYMER AND CELLULOSIC FIBRES, AND COMPOUNDED MATERIALS OBTAINED THEREWITH



## (57) Abstract

The present invention relates to a process for continuously manufacturing composites of polymer and cellulosic fibres, comprising the steps of: a) feeding a polymer upstream into an extruder; b) melting and mixing the polymer in a zone (A) of the extruder; wherein zone (A) comprises at least one positive transportation screw element; c) feeding cellulosic fibres into the extruder in a zone (B) of the extruder, which zone (B) is located downstream of zone (A); d) transporting the mixture of polymer and cellulosic fibre obtained in zone (B) through a degassing zone (C), which zone (C) is located downstream of zone (B), wherein zone (C) comprises at least one positive transportation screw element and e) transporting the mixture obtained in zone (C) through a pressure building zone (D) of the extruder, which zone (D) is located downstream of zone (C), wherein zone (D) comprises at least one positive transportation screw element; f) releasing the mixture obtained in zone (D) into a die, characterised in that zone (B) comprises at least one positive transportation screw element, at least one kneading section and at least one negative transportation screw element such that in zone (B) of the extruder the cellulosic fibres are fibrillated to obtain cellulosic fibres with an aspect ratio as high as possible, while simultaneously mixing the cellulosic fibres with the melted polymer. This process results in a compounded material comprising polymer and cellulosic fibres, wherein the cellulosic fibres are elementary fibres and have a high aspect ratio. As a result the compounded material has excellent mechanical properties.

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**Process for continuously manufacturing composites of polymer and cellulosic fibres, and compounded materials obtained therewith**

- 5 The present invention relates to a process for continuously manufacturing composites of polymer and cellulosic fibres and to the compounded material obtained therewith. It also relates to an extruder to be used in this process.

It is known to produce fibre reinforced plastics. For instance GB 1,151,964 describes a  
10 method for obtaining a plastics material reinforced with brittle fibres such as glass fibres. According to this method a brittle fibre substance is supplied as a continuous strand to the other components of the mixture in such a manner that the fibre breaks to a predetermined length. The apparatus used for this process comprises several mixing and kneading members which are not specified.

15

Lately, the focus on reinforcing fibres is shifting from glass fibres to certain kinds of cellulosic fibres which have outstanding intrinsic mechanical properties. These have the potential to compete with glass fibres as reinforcing agents in plastics. The specific strength of these agrofibre is 50 to 80 percent of glass fibres, whereas the specific  
20 modulus can exceed that of glass fibres. Supplementary benefits include low cost, low density, renewability and (bio)degradability. In addition, they are less abrasive during processing with thermoplastics and do not expose operators to potential safety or health problems.

- 25 A major disadvantage of cellulosic fibres is the limited temperature at which they should be processed without losing their additional mechanical properties. Further, it has been proven difficult to obtain a homogeneous mixture of polymer and fibre. This is mainly due to the non-polar polymeric surface versus the highly polar fibre surface which prevents satisfactory fibre/polymer intertwining.

30

EP 426,619 describes a method of producing panels from a thermoplastic polymer and a thermosensitive filler by means of an extruder having at least three feedingly effective helical extrusion sections and at least two non-feeding kneading sections. The extruder

thus comprises at least two kneading zones and at least three extrusion zones. The filler is preferably fed into the second extrusion zone.

When cellulosic fibres are used it is important that during the extrusion process these  
5 fibres obtain and maintain a high aspect ratio so as to obtain a compounded material with mechanical properties comparable with those of materials containing glass fibres. This means that the diameter should be as small as possible, preferably so called elementary fibres are used. Further the length of the fibres should be as large as possible.

10 Obtaining such a high aspect ratio of the fibres in the final product has up to now not been achieved. The problem with extrusion processes according to the state of the art is that the high shear forces in the extruder often result not only in a smaller diameter of the fibres but also in a smaller length of the fibres. This reduces the strength properties of the compounded material considerably, relative to the glass fibre reinforced materials.

15

Accordingly, a substantial need exists for a continuous process for the production of polymer/cellulosic fibre composites with substantially improved mechanical properties, i.e. its rigidity and its strength. In addition, these resulting property improvements should be valid for multiple fibre sources at broad polymer processing temperature ranges and  
20 independent from the polymeric melt behaviour.

The above objects are achieved by the present invention. The present invention relates to a process for continuously manufacturing composites of polymer and cellulosic fibres, comprising the steps of:

- 25 a) feeding a polymer upstream into an extruder;  
b) melting and mixing the polymer in a zone (A) of the extruder; wherein zone (A) comprises at least one positive transportation screw element,  
c) feeding cellulosic fibres into the extruder in a zone (B) of the extruder, which zone (B) is located downstream of zone (A);  
30 d) transporting the mixture of polymer and cellulosic fibre obtained in zone (B) through a degassing zone (C), which zone (C) is located downstream of zone (B), wherein zone (C) comprises at least one positive transportation screw element and  
e) transporting the mixture obtained in zone (C) through a pressure building zone (D)

of the extruder, which zone (D) is located downstream of zone (C), wherein zone (D) comprises at least one positive transportation screw element.

f) releasing the mixture obtained in zone (D) into a die, characterised in that

5 zone (B) comprises at least one positive transportation screw element, at least one kneading section and at least one negative transportation screw element such that in zone (B) of the extruder the cellulosic fibres are fibrillated to obtain cellulosic fibres with an aspect ratio as high as possible, while simultaneously mixing the cellulosic fibres with the melted polymer.

10

The design of this process is such that during the continuous mixing the cellulosic fibres are opened up to elementary fibres (fibrillation) with a high aspect ratio, which are homogeneously distributed in the polymeric melt. The process results in a compounded material with improved rigidity and strength.

15

The present invention also relates to an extruder which can be used to carry out the process. The present invention comprises all extruders with two separate feeding ports and a degassing port. The preferred extruder for performing the process of the present invention is a corotating twin-screw extruder. An example of such an extruder is a  
20 Berstorff ZE corotating twin-screw extruder with a length to diameter ratio varying from 35 to 40.

As indicated above, the extruder comprises four zones: a zone (A) where a polymer fed to the extruder is melted and mixed; a zone (B) where cellulosic fibres are fed to the  
25 extruder, fibrillated to elementary fibres and simultaneously mixed with the polymer; a zone (C) where the mixture of polymer and cellulosic fibre obtained in zone (B) is degassed and a zone (D) where pressure is built up.

According to the invention zone (A), which is the polymer melting and mixing zone,  
30 comprises at least one positive transportation screw element. Preferably zone (A) further comprises at least one kneading section and at least one negative transportation screw element. Preferably, zone (A) begins at a distance of  $20 \times D$  calculated from the beginning of the die, wherein D stands for the diameter of the extrusion screw. Generally zone (A)

ends at a distance of  $38 \times D$ .

In this application the location of the zones is calculated from the beginning of the die, i.e. from the end of the extruder. This is contrary to general practice in which these distances  
5 are calculated from the beginning of the extruder. This is done because according to the invention it is important that the feeding port for the fibres is located as close as possible to the end of the extruder.

Zone (A) can further be divided into four temperature zones. Zone (A1) defines the  
10 feeding of the polymer. In this zone, which is generally located between  $34 \times D$  and  $38 \times D$ , the polymeric material is fed to the extruder. A conventional feeder in combination with a hopper are generally used for this purpose. After being fed to the extruder, the material is transported to zone (A2).

15 In zone (A2), which is generally located between  $30 \times D$  and  $34 \times D$ , the polymeric material starts to melt, predominantly by shear forces. From (A2) the material is transported to (A3), which is located between  $24 \times D$  and  $30 \times D$ . From this zone the polymer is transported to zone (A4) located between  $24 \times D$  and  $20 \times D$ . Both zone (A3) and (A4) serve to further melt and mix the polymer.

20

Zone (B), which is the fibre fibrillation and mixing zone, comprises at least one positive transportation screw element, at least one kneading section, preferably at least two kneading sections, and at least one negative transportation screw element. Zone (B) is preferably located at a distance between  $8 \times D$  and  $20 \times D$ . The at least one kneading  
25 section of zone (B) is preferably located at a distance between  $10 \times D$  and  $13 \times D$ .

Zone (B) comprises two temperature zones: (B1) and (B2). In zone (B1) the predried cellulosic fibres are continuously and gravimetrically fed in a conventional manner, for instance from a feeder to a hopper and further to the extruder. The position of this zone is  
30 between  $14 \times D$  and  $20 \times D$ . Preferably the fibres are fed at  $16 \times D$ . The amount of fibres fed is such that the weight ratio of the fibres in the final compounded material is controlled to a certain value. In addition the fibres start to be distributed in this zone.

In zone (B2) the fibres are fibrillated to elementary fibres. In addition the fibres are distributed homogeneously through the polymer matrix. If more than one positive transportation screw element is present in zone (B2), these elements preferably have a decreasing pitch in the flow direction of the polymer and fibre mixture. This zone is  
5 located between  $8 \times D$  and  $14 \times D$ .

Zone (C), the degassing zone, comprises at least one positive transportation screw element. In this zone water and other thermally unstable components are removed from the compounded mixture. For this purpose a conventional degassing port is present which  
10 is connected to a vacuum pump. At the end of this zone when the volatile substances have substantially been removed, binding of the fibres and matrix begins.

Zone (D) the pressure building zone, comprises at least one positive transportation screw element. Preferably this zone comprises at least two positive transportation elements. In  
15 that case these elements have a decreasing pitch in the direction of the die. This results in an increase in pressure necessary to press the compounded material through the die.

In this zone further homogeneous distribution of the fibres into the matrix and compaction of the compounded material is accomplished resulting in the penetration of polymeric  
20 material into the surface pores and microcracks of the cellulosic fibres. During this process both mechanical interlocking and chemical coupling of matrix and fibres are further increased resulting in optimised fibre/matrix interaction. Zone (D) comprises one temperature zone.

25 The temperatures of the different temperature zones described above depend upon the kind of polymer used. If the polymer is polyethylene, polypropylene or polystyrene, the following temperature profile may be applied.

Zone	Temperature (°C)
A4	25 to 160
A2	165 to 185
A3	190 to 210
A4	190 to 210
B1	185 to 205
B2	185 to 205
C	180 to 200
D	185 to 205

The thermoplastic polymers that can be applied in the invention comprise "commodity" plastics like low density polyethylene, high density polyethylene, poly(ethylene-copropylene), polypropylene (homopolymer and copolymer) and polystyrene  
 5 (homopolymer, copolymer and terpolymers). In addition engineering plastics can be applied. Besides the virgin polymeric material, the recycled grades of the above-mentioned plastics are also applicable in the present invention.

A general definition of cellulosic fibres for the purpose of the present invention is 'any  
 10 fibres were the main constituents are of plant tissue and whose main component consist of  $\alpha$ -cellulose'. Preferably annual plant fibres or bast fibres, like flax, hemp, jute and kenaf are used. According to a different embodiment paper fibres such as recycled fibres from newspaper are used. It is also possible to use a combination of different types of fibres such as paper fibres and bast fibres. Annual growth plant fibres can, by means of their  
 15 intrinsic mechanical properties, generally compete with glass fibres.

Generally the raw cellulosic fibre bundles are between one and five millimetres in diameter. After being fibrillated during the compounding process, the cellulosic fibre diameter generally varies from ten to hundred micrometers, whereas the fibre aspect ratio  
 20 varies from 7 to 100.



The present process is especially suitable for a feed consisting of raw cellulosic fibres which is very economical. However, it is also possible to feed elementary fibres to the extruder. The present process is then advantageous because the elementary cellulosic  
5 fibres retain their high aspect ratio.

The amount of fibres fed to the extruders is such that in the final compounded material the fibre content is 5 wt.% to 50 wt.% based on the weight of the compounded material, preferably 30 wt.% to 40 wt.%, most preferably 40 wt.%.

10

According to the present invention it is preferred to add a coupling agent to the polymeric material. With a coupling agent is meant a polymer which can be mixed with the polymer matrix and which can be chemically coupled onto the fibres. The preferred polymer to coupling agent ratio for a polyolefinic matrix (e.g. polyethylene, polypropylene) is 70 to 6,  
15 with a most preferred ratio of 8 to 16, based on weight. For the polystyrene matrix the preferred polymer to coupling agent ratio is 700 to 60, 450 to 550 being most preferred.

The preferred coupling agent for polyethylene is maleic anhydride grafted polyethylene copolymer. The preferred coupling agent for polypropylene is maleic anhydride grafted  
20 polypropylene copolymer. The preferred coupling agent for polystyrene is maleic anhydride grafted polystyrene copolymer.

The coupling agent is preferably dry mixed with the polymer before being introduced into the extruder. For this purpose any conventional blender may be used. The mixture thus  
25 obtained is fed to the feeder as mentioned above. The polymer and coupling agent are homogeneously mixed in zone (A) of the extruder.

Other additives which can be added to the polymer comprise conventional additives such as pigments, antioxidants, fillers and flame retardants. Examples of fillers that may be  
30 used are talc, calcium carbonate and carbon black.

The mechanical properties of the compounded material according to the present invention compared with the properties of the polymer matrix are shown below.

Material	Stiffness [GPa]	Strength [MPa]
PP	1.2	41
PP/cellulose fibre	5.8	95
LDPE	0.1	6
LDPE/cellulose fibre	1.6	28
HDPE	0.9	20
HDPE/cellulose fibre	3.5	47
PS	1.9	40
PS/cellulose fibre	6.0	65

PP = polypropylene

LDPE = low density polyethylene

5 HDPE = high density polyethylene

PS = polystyrene

% cellulose fibre = 40 % by weight

The compounded material exiting the die may be granulated before being further  
 10 processed. These granules can be formed into articles by means of thermoforming  
 processing techniques such as injection molding and compression molding. It is also  
 possible to directly mould the compounded material obtained into sheets, tubes, profiles,  
 etc. The articles obtained from the compounded material may serve to replace wood,  
 plastic and alternatively filled- or reinforced composite alternatives.

15

The invention shall now be described in more detail by means of the annexed drawing.  
 According to the present process a polymer is fed to the extruder at feeding port (21),  
 (zone A1). Before feeding, the polymer is preferably mixed with a coupling agent in a  
 blender (not shown), added to a feeder (not shown) and the dry mixture is then  
 20 gravimetrically fed from the feeder through a hopper (not shown) into the extruder (20).

The polymeric material starts to melt, predominantly by shear forces, as it is transported through zone (A2). In zone (A3) the material is further melted; in addition both polymer and coupling agent are homogeneously mixed, whilst potential temperature differences  
5 have vanished to a large extent. Distributive mixing of the coupling agent in the polymeric matrix is further optimised in zone (A4).

In zone (B1) predried cellulosic fibres are continuously and gravimetrically fed at feeding port (22) from a feeder to a hopper (not shown). In this zone the cellulosic fibres are  
10 introduced into the polymeric melt. In zone (B2), the fibres are virtually fibrillated to elementary fibres in the kneading section. In addition the fibres start to be distributed in this zone.

Following the transporting screw elements with decreasing pitch, resulting in increasing  
15 pressure in zone (B2), the fibres are further homogeneously distributed in the polymeric matrix. In zone (C) water and other thermally unstable components are removed from the compounded mixture in degassing port (23), to a vacuum pump (not shown). With the volatiles being removed chemical coupling of fibres and matrix preferably starts.

20 In zone (D) homogeneous distribution of the fibres into the matrix and compaction of the compounded material is accomplished resulting in the penetration of (maleic anhydride grafted) polymeric material into the surface pores and microcracks of the cellulosic fibres. During this process both mechanical interlocking and chemical coupling of matrix and fibres are further increased resulting in optimised fibre/matrix interaction.

25

Since the cellulosic fibres are introduced in to the polymeric melt as late as possible, the fibrillated material is the least affected by friction and heat. As a result the fibre aspect ratio remains as high as possible under the described processing conditions, which leads (in combination with optimised chemical and mechanical fibre/matrix coupling) to  
30 substantially improved material stiffness and strength properties.

### Examples

Several screw configurations were used to prepare a mixture of polymer and fibre. The

extruder was a Berstorff ZE corotating twin-screw extruder. For each example the extruder compounding conditions were, unless indicated otherwise:

screw speed	200 rpm
melt temperature	195 °C
5 matrix material	polypropylene homopolymer MFI <sub>230,2.16</sub> = 12 g/10 min.
fibre content	30 wt.%
fibre type	kenaf (except example 5)

For all examples the fibre length and the percentage of fibres still present as a bundle of  
 10 the agrofibres in the matrix material was determined. The fibre dimension is an indication of the rigidity and strength of the final material. In order to determine fibre length the agrofibres were extracted from extruder compounded PP/fibre granules using soxhlet extraction with decaline as the solvent. Agrofibre-length measurements were performed at a Kajaani FS-200 following Tappi T271 pm-91.

15

#### Example 1 (comparative)

For this example a screw configuration according to the following table was used:

Screw Type	SW	SW	SW	KB	KB	RSE	SW	SW	SW
Amount	6	1	1	1	1	1	14	1	3
Length (mm)	60	40	30	50	30	20	60	40	30
Transport	+	+	+	+	-	-	+	+	+
Pitch (mm)	60	40	30	100	60	40	60	40	30

20

In this table SW stands for Self Wiping, KB stands for Kneading Block and RSE stands for Reverse Screw Element.

The fibre lengths could not be measured due to a stagnating extruder. This was caused  
 25 by large numbers of unopened fibre bundles, which constipated the extruder die.

**Example 2 ("heavy" fibre mixing and opening)**

For this example a screw configuration according to the following table was used:

Screw Type	SW	SW	SW	KB	KB	RSE	SW	KB	KB
Amount	6	1	1	1	1	1	7	1	1
Length (mm)	60	40	30	50	30	20	60	30	30
Transport	+	+	+	+	-	-	+	+	+
Pitch (mm)	60	40	30	100	60	40	60	60	60

5 [continued..]

Screw Type	KB	RSE	SW	SW	SW	KB	KB	RSE	SW	SW
Amount	1	1	3	1	1	1	1	1	1	3
Length (mm)	30	20	60	30	20	30	30	20	40	30
Transport	-	-	+	+	+	+	-	-	+	+
Pitch (mm)	60	40	60	20	20	60	60	40	40	30

The results of this example are indicated in Table 1.

10

**Example 3 (optimal fibre mixing and opening)**

For this example a screw configuration according to the following table was used:

Screw type	SW	SW	SW	KB	KB	RSE	SW	KB	KB	RSE	SW	SW	SW	KB	SW	SW	SW
Amount	6	1	1	1	1	1	7	1	1	1	1	1	1	1	3	1	3
Length (mm)	60	40	30	50	30	20	60	30	30	20	60	40	30	30	60	40	30
Transport	+	+	+	+	-	-	+	+	-	-	+	+	+	+	+	+	+
Pitch (mm)	60	40	30	100	60	40	60	60	60	40	60	40	30	60	60	40	30

15

The results of this example are indicated in Table 1.

**Example 4 (batch kneader)**

The compound preparation was performed by batch kneading on a HAAKE Rheomix  
 20 3000 equipped with roller rotors. The Rheomix was operated at 185°C and 100 RPM.  
 The PP granulate was added first and kneaded for 2 minutes, followed by the

introduction of the kenaf fibres. This which was kneaded for 7 minutes, after which a composite material with homogeneously dispersed fibres was produced.

#### Example 5

- 5 Example 3 was repeated with the exception that flax fibres were used instead of kenaf fibres. The results are indicated in Table 1.

Table 1

Example	Arithmic average [mm]	Length weighted average [mm]	number of bundled fibres [%]
1	-	-	<i>innumerable</i>
2	1.17	1.63	19
3	1.51	1.93	22
4	0.28	0.44	1
5	0.63	1.43	<i>not measured</i>

### Claims

1. Process for continuously manufacturing composites of polymer and cellulosic  
5 fibres, comprising the steps of:
- a) feeding a polymer upstream into an extruder;
  - b) melting and mixing the polymer in a zone (A) of the extruder; wherein zone (A)  
comprises at least one positive transportation screw element,
  - c) feeding cellulosic fibres into the extruder in a zone (B) of the extruder, which zone  
10 (B) is located downstream of zone (A);
  - d) transporting the mixture of polymer and cellulosic fibre obtained in zone (B)  
through a degassing zone (C), which zone (C) is located downstream of zone (B),  
wherein zone (C) comprises at least one positive transportation screw element and
  - e) transporting the mixture obtained in zone (C) through a pressure building zone (D)  
15 of the extruder, which zone (D) is located downstream of zone (C), wherein zone  
(D) comprises at least one positive transportation screw element.
  - f) releasing the mixture obtained in zone (D) into a die,  
characterised in that  
zone (B) comprises at least one positive transportation screw element, at least one  
20 kneading section and at least one negative transportation screw element such that in zone  
(B) of the extruder the cellulosic fibres are fibrillated to obtain cellulosic fibres with an  
aspect ratio as high as possible, while simultaneously mixing the cellulosic fibres with the  
melted polymer.
- 25 2. Process according to claim 1, wherein zone (A) further comprises a kneading  
section and at least one negative transportation screw element
3. Process according to claim 1 or 2, wherein zone (B) comprises at least two  
kneading sections.
- 30 4. Process according to any of claims 1 to 3, wherein zone (D) comprises at least  
two positive transportation screw elements with decreasing pitch towards the die.

5. Process according to any one of claims 1 to 4, wherein the cellulosic fibres are bast fibres and/or paper fibres.

6. Process according to any one of claims 1 to 5, wherein zone (B) is located at a distance between  $8 \times D$  and  $20 \times D$  calculated from the beginning of the die,  $D$  being the diameter of the screw.

7. Process according to any one of claims 1 to 6, wherein the fibres are fed into the extruder at a distance between  $14 \times D$  and  $20 \times D$ , preferably  $16 \times D$ , calculated from the beginning of the die.

8. Process according to any one of claims 1 to 7, wherein the at least one kneading section of zone (B) is located at a distance between  $10 \times D$  and  $13 \times D$ , calculated from the beginning of the die.

9. Process according to any one of claims 1 to 8, wherein zone (C) is located at a distance between  $4 \times D$  and  $8 \times D$  calculated from the beginning of the die.

10. Process according to any one of claims 1 to 9, wherein zone (D) is located at a distance between  $0 \times D$  and  $4 \times D$  calculated from the beginning of the die.

11. Process according to any one of claims 1 to 10, wherein a coupling agent is mixed with the polymer before feeding the polymer to the extruder.

12. Process according to any of claims 1 to 11, wherein the fibres are gravimetrically fed in such an amount as to obtain a final compounded material containing 5 to 50 wt.%, preferably 30 to 40 wt.% fibres on the basis of the weight of the compounded material.

13. Process according to any of claims 1 to 12, wherein the extruder is a corotating twin-screw extruder.

14. Compounded material obtainable by the process according to any of claims 1 to 13.



15. Extruder for continuously manufacturing composites of polymer and cellulosic fibres connected to means for feeding a polymer, means for feeding cellulosic fibres and means for degassing the extruder, comprising:

5       a zone (A) where a polymer fed to the extruder is melted and mixed, comprising at least one positive transportation screw element,

          a zone (B) where cellulosic fibres are fed to the extruder, fibrillated and simultaneously mixed with the polymer, comprising at least one positive transportation screw element, at least one kneading section and at least one negative transportation screw  
10   element,

          a zone (C) where the mixture of polymer and cellulosic fibre obtained in zone (B) is degassed, comprising at least one positive transportation screw element,

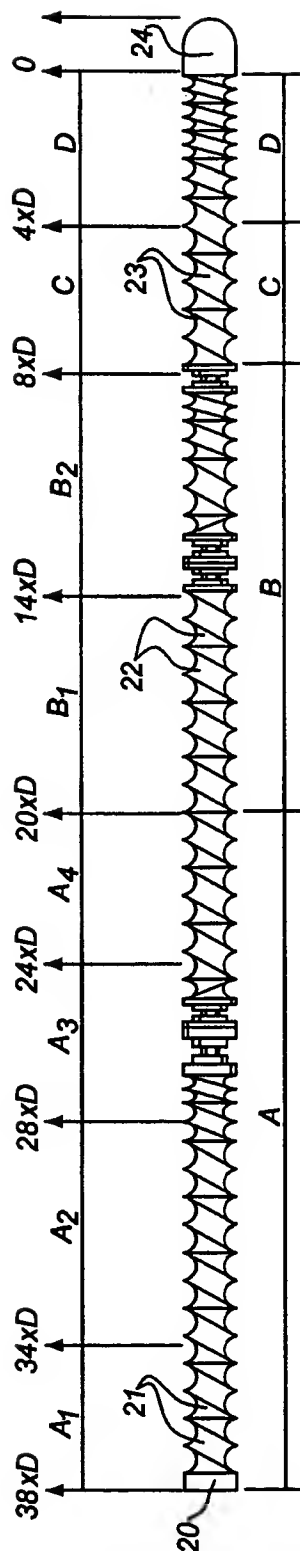
          a zone (D) where pressure is built up, comprising at least one positive transportation screw element and

15       a die.

16. Extruder according to claim 15, wherein zone (A) further comprises a kneading section and at least one negative transportation screw element, zone (B) comprises at least two kneading sections and zone (D) comprises at least two positive transportation screw  
20   elements with decreasing pitch towards the die.

17. Extruder according to claim 15 or 16, which is a corotating twin-screw extruder.

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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/NL 99/00282

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 B29C47/00 B29C47/10 B29C47/40 B27N3/28

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B29C B27N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 426 619 A (ICMA IND COSTR. MAC AFFINI) 8 May 1991 see abstract see column 1, line 45 - column 2, line 14 see column 3, line 53 - column 4, line 12 see column 5, line 24 - column 6, line 16 see column 8, line 2 - line 23 see column 8, line 54 - column 9, line 27 see claims; figures 1-6	1-17
X	GB 1 151 964 A (WERNER & PFLEIDERER) 14 May 1969 see page 2, line 92 - page 3, line 14 see claim 1; figures	15-17
A	---	1-14
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search

1 July 1999

Date of mailing of the international search report

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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/NL 99/00282

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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A	EP 0 118 847 A (HENCH AUTOMATIK APP MASCH) 19 September 1984 see page 8, line 13 - page 9, line 32 see claims; figure 2 ---	1-17
A	PATENT ABSTRACTS OF JAPAN vol. 007, no. 215 (M-244), 22 September 1983 & JP 58 110233 A (TOSHIBA KIKAI KK), 30 June 1983 see abstract ---	15-17
A	GB 2 140 314 A (CHISSO CORP) 28 November 1984 see abstract see claims 1-9; figures see page 2, line 59 - page 3, line 25 ---	1
A	WO 97 30838 A (FORMTECH ENTERPRISES INC) 28 August 1997 see the whole document -----	1

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Information on patent family members

International Application No

PCT/NL 99/00282

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